AN ION SOURCE FOR THE HVEE ^{14}C ISOTOPE RATIO MASS SPECTROMETER FOR BIOMEDICAL APPLICATIONS

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ABSTRACT. During the past two decades, accelerator mass spectrometry (AMS) has allowed major developments in many areas of geosciences and archaeology. In the near future, AMS should realize a similar potential in the field of biomedical research, leading ultimately to clinical applications. For such applications, the required instrument differs significantly from that presently used in the field of ^{14}C dating. Whereas the needed accuracy and sensitivity is more than an order of magnitude less demanding than that for present state-of-the-art ^{14}C instrumentation, the widespread acceptance of ^{14}C AMS in biomedical research will require AMS spectrometers that are small, simple to operate and capable of handling CO_2 samples. In order to satisfy these demands, HVEE has developed a compact ^{14}C AMS spectrometer dedicated to biomedical research. The instrument consists of a compact accelerator with a footprint of 2.25 × 1.25 m and an ion source that features direct CO_2 acceptance and optimal user friendliness. Having previously described the layout and design of the accelerator, we here discuss progress on the accelerator and present the design and first results of the CO_2 ion source.

INTRODUCTION

In the past two decades, AMS has developed into a mature scientific tool for a variety of applications in the geosciences and archaeology. The usefulness of AMS has been recognized in other disciplines as well, notably biomedical research (Davis *et al.* 1990; Davis 1994), as demonstrated by an exponential increase in the number of contributions in this field at recent AMS conferences in Canberra-Sydney (Fifield *et al.* 1994) and Tucson (Jull, Beck and Burr 1997).

The required instrumentation for biomedical research differs significantly from that presently used in the field of ^{14}C dating. The use of AMS instrumentation within the biomedical community is presently handicapped by the size of existing systems and their need for expensive support personnel; widespread acceptance will require small AMS spectrometers that need little or no maintenance and that are easy to operate. Size reduction of the instrument can be achieved by sacrificing sensitivity and accuracy, since these specifications are at least one order of magnitude less demanding in biomedical research. Throughput requirements are an order of magnitude higher, however.

The present AMS technique relies on a laborious graphitization procedure that is unknown to the biomedical community. The introduction of AMS instrumentation to this community would be eased by the availability of ion sources that directly accept CO_2, enabling direct interfacing with a CHN analyzer or a gas chromatograph.

HVEE has developed a compact AMS spectrometer dedicated to biomedical research that overcomes the shortcomings of the presently available AMS instruments. With a footprint of 2.25 × 1.25 m, it is easy to operate and capable of analyzing CO_2 samples. Previously (Mous *et al.* 1997) we reported on the design and layout of the instrument, whose accuracy and throughput have yet to be fully characterized. Here we discuss progress on the system and present the development and first results of a compact cesium-type sputter source that directly accepts CO_2.

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[1] These authors were exclusively involved in the design and development of the CO_2 ion source.

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THE CO_2 ION SOURCE

The design of the CO_2 ion source results from cooperation between HVEE and Oxford University. Its design is based on that of the OXGIS source developed at Oxford (Bronk 1990), with improvements seeking to minimize memory effects and optimize user friendliness. The operation characteristics of the ion source are reflected in the following features:

- acceptance of both graphite samples and CO_2 samples admitted from ground potential;
- an easily exchanged 50-sample carousel;
- source embodiment at ground potential ensuring easy and safe operation;
- almost complete absence of radiation hazard as a result of a stainless-steel source embodiment;
- optimal vacuum conditions for low memory effects;
- easy, quick source maintenance *via* sideways disassembly of the source-head.

A schematic layout of the Cs sputter-type ion source is shown in Figure 1. Its principle of operation is well understood and described extensively in the literature (*e.g.*, Alton 1990). In this section we concentrate on the technical aspects of the new source design.

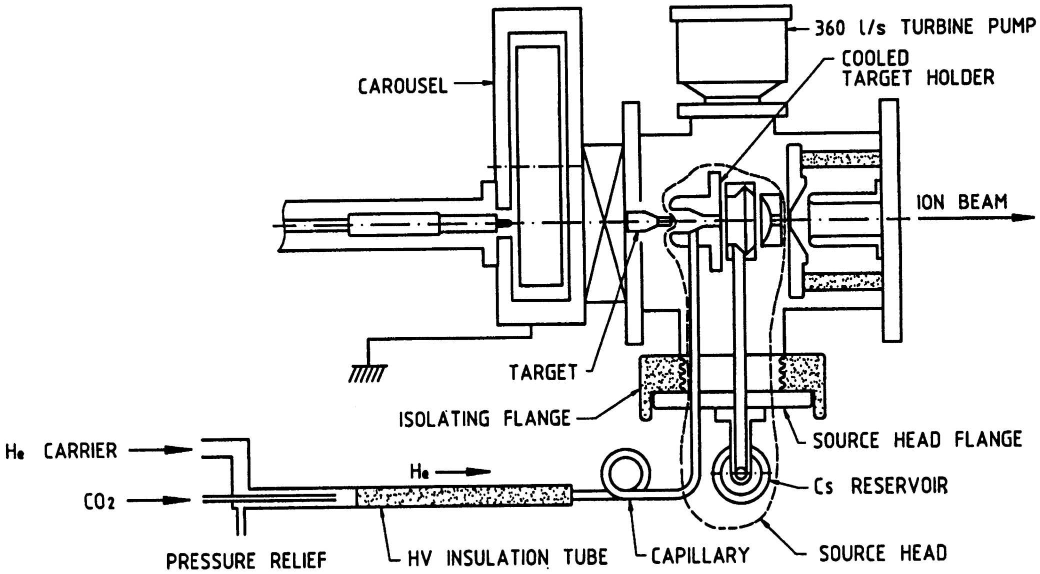

Fig. 1. Schematic layout of the CO_2 ion source

The stainless-steel source embodiment, including carousel, is at ground potential during operation, avoiding the necessity of a large isolation cage, thereby reducing the space requirements of the total assembly. Isolation of the extraction and target voltage is achieved by insulators within the source embodiment (Fig. 1). All high-voltage connections to the source, including the Cs reservoir, the CO_2 feed tube and the cooling lines, are brought together on the source-head insulation flange, keeping the space needed for external high-voltage isolation to a minimum. The source-head (Fig. 2) can be pulled sideways from the source embodiment, making maintenance easy and quick.

A 360 L s^{-1} turbomolecular pump is directly mounted on the source embodiment, just above the source-head, providing optimal pumping speed in the source area. This fulfills an important condi-

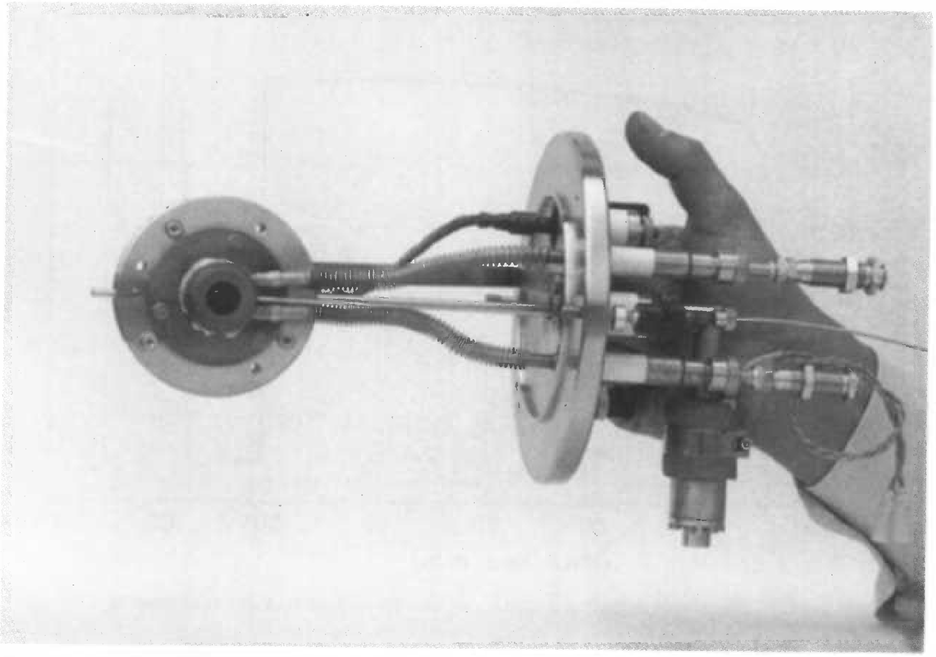

Fig. 2. Photograph of the source-head

tion for low memory in the case of CO_2 targets, which is of paramount importance for biomedical applications, where the ^{14}C content of a sample can vary by many orders of magnitude.

The sample carousel can accommodate up to 50 samples and is isolated from the source by a 100-mm diameter gate-valve, ensuring good vacuum conditions at the carousel at all times. A pneumatically operated cylinder transports the sample under investigation into the ion source.

The source accepts graphite as well as CO_2 samples. Solid graphite sample material is pressed in a target holder in a similar manner as is done in most modern AMS laboratories. CO_2 samples can be admitted into the ion source from ground potential. A He carrier of *ca.* 1 sccm transports the CO_2 *via* an isolation tube and a stainless-steel capillary into the ion source (Fig. 1). Here, the CO_2 is absorbed on the surface of a titanium target, where it is sputtered and ionized to form a negative carbon beam. During gas admission, the vacuum level in the source is maintained at *ca.* 1×10^{-5} mbar.

OPERATION AND FIRST RESULTS

The source is operated at a target potential of 6 kV and 19 kV extraction, giving a 25 keV ion energy. Analyzed ^{12}C currents from graphite samples were measured at between 50 and 100 μA, with moderate Cs supply into the ion source. No attempts have been made to push the source to its limits, but currents well in excess of 100 μA should be achievable. Figure 3 shows the analyzed $^{12}C^-$ current for different graphite samples as a function of time. After target insertion, the ^{12}C current rises quickly, stabilizing within *ca.* 30 s. Furthermore, it can be seen from the figure that the sample transfer time (for neighboring samples) is *ca.* 7 s. During sample exchange ($t = \sim 55$ s in Fig. 3), a few μA of $^{12}C^-$ current is still present. This current probably results from the distorted ion optics of the sputtering Cs beam due to the absence of the target in the source and does not reflect actual source background.

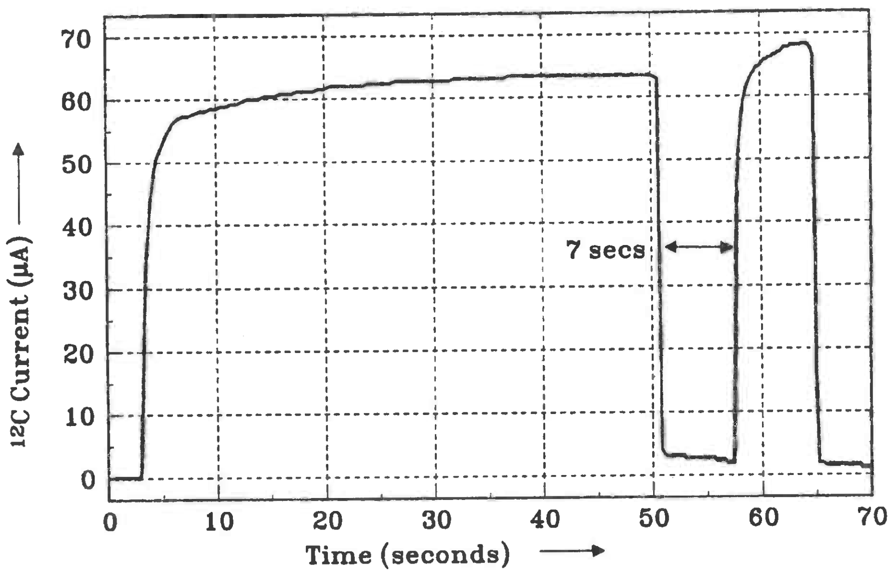

Fig. 3. Analyzed $^{12}C^-$ ion source current as a function of time for two different graphite samples

The Cs spot size on the target was measured at *ca.* 0.8 mm in diameter, which corresponds reasonably well to the design value of 0.6 mm. The emittance of the source for a 50 μA ^{12}C beam was measured at *ca.* 6 mm mrad $\text{MeV}^{-.5}$.

Exchanging of the carousel, which houses 50 targets, is easy; pump-down takes *ca.* 5 min before the next analysis can be started.

The source was briefly tested with CO_2 samples, using a He carrier of *ca.* 1 sccm to transport the CO_2 gas from ground potential into the source (Fig. 1). Analyzed ^{12}C currents of 5–10 μA were achieved, which is *ca.* one order of magnitude less than currents from graphite samples. Further tests in progress seek to optimize source output and to quantify the memory effects of the source in the case of CO_2 samples.

PROGRESS ON THE ^{14}C AMS SPECTROMETER

Figure 4 shows a schematic diagram of the HVEE biomedical ^{14}C AMS spectrometer. Negative ions are extracted from the ion source and mass analyzed by a small 90° permanent magnet. At the exit of the magnet, ^{12}C ions are collected in a Faraday cup for normalization; ions with a mass of 14 proceed to pass a 90° electrostatic analyzer (ESA), the tandem accelerator with a 180° ESA in its terminal and a magnetic deflector before they are collected in a particle detector. A detailed description, together with the principles of background reduction of the instrument, is given elsewhere (Mous *et al.* 1997).

Since the early tests on the system, various improvements have been made to optimize the beam optics. A remotely controlled power supply in the high-voltage terminal for the adjustment of the ESA and a gridded lens following the high-energy (HE) tube were incorporated.

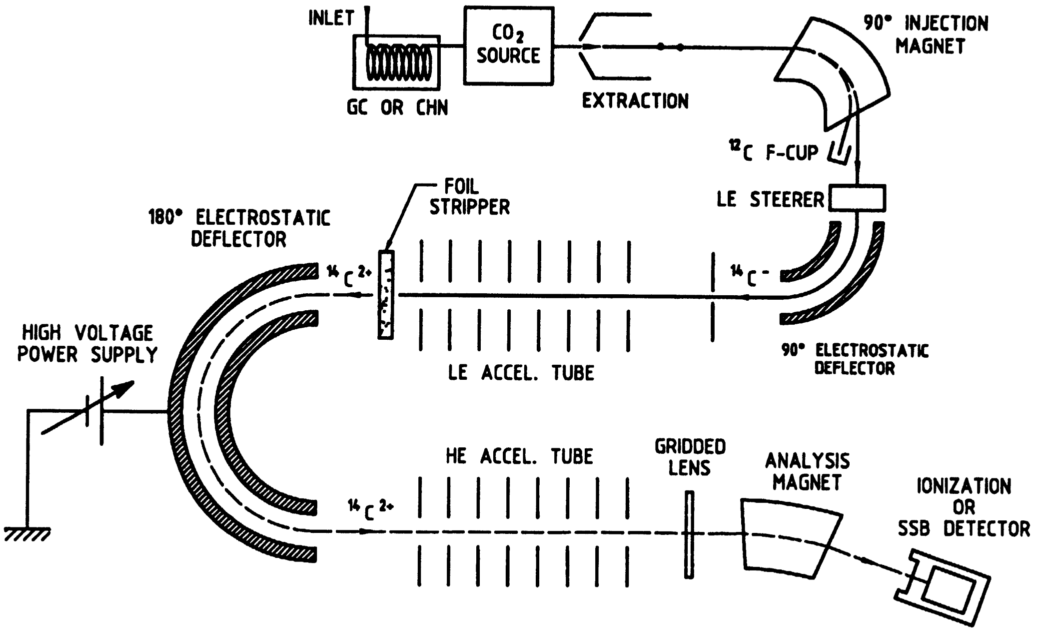

Fig. 4. Schematic layout of the HVEE ^{14}C AMS spectrometer

To evaluate the ion optical quality of the accelerator, the $^{13}C^{2+}$ current in the HE Faraday cup located just in front of the detector was measured during a scan of the low energy (LE) steerer and the 180° ESA in the terminal. Figure 5 presents the results. Both traces show a flat-top tuned transmission, indicating that the beam is transported through the system without loss. Furthermore, the figure indicates the usefulness of the LE aperture during tuning of the system.

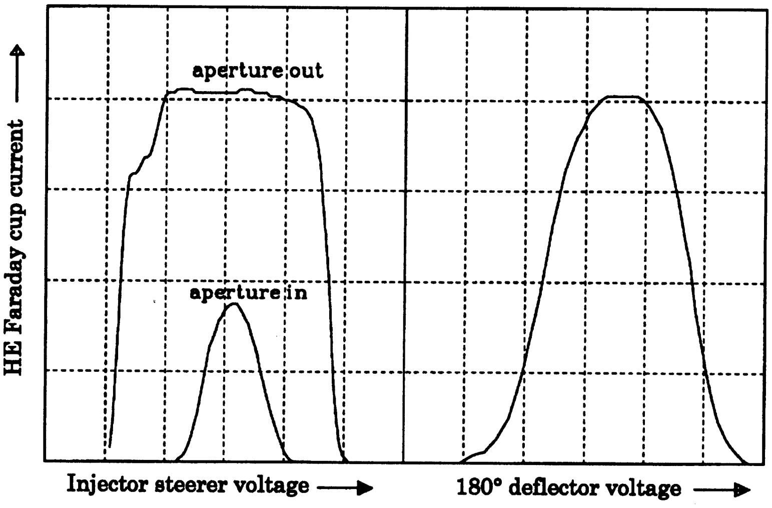

Fig. 5. HE Faraday cup current as a function of injector steerer voltage and 180° electrostatic analyzer voltage

CONCLUSION

The cooperation between HVEE and Oxford University has resulted in a unique, compact ion source that accepts both graphite samples and CO_2 samples from ground potential. The source will be applied on the HVEE ^{14}C AMS spectrometer dedicated to biomedical research. Early tests have showed that analyzed $^{12}C^-$ currents of 50–100 μA are easily extracted from graphite samples, whereas 5–10 μA is obtained from CO_2 samples.

Beam transmission of the HVEE ^{14}C AMS spectrometer is flat-top tuned, which guarantees that accurate ^{14}C analysis can be achieved.

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